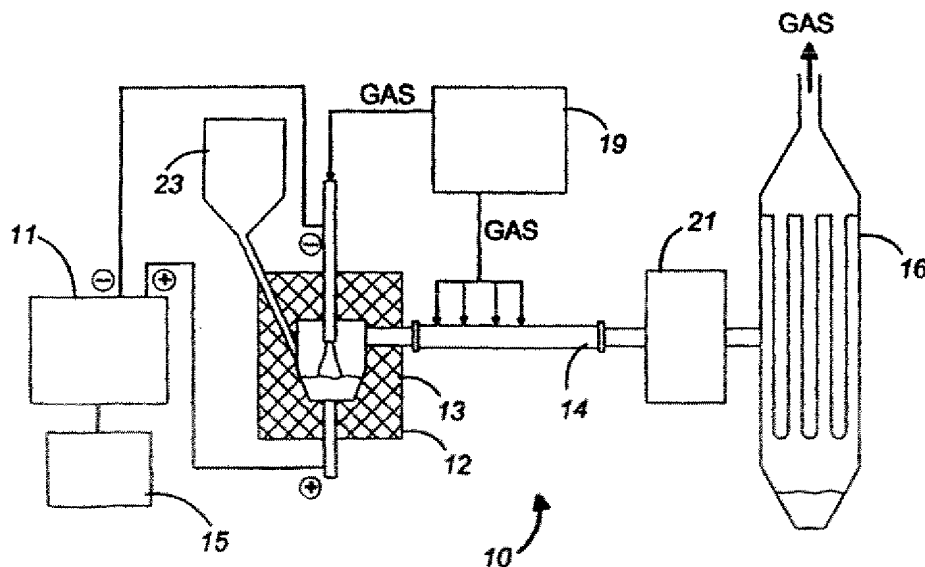# PCT

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(54) Title: METHOD AND TRANSFERRED ARC PLASMA SYSTEM FOR PRODUCTION OF FINE AND ULTRAFINE POWDERS

(57) Abstract

The present invention is concerned with a method for the production of fine and ultrafine powders of various materials such as metals, alloys, ceramics, composites etc., through a transferred arc plasma system. The method comprises vaporizing or decomposing the material in the plasma reactor, condensing the vapor in a quench tube comprising two sections, the first one for indirectly cooling or heating the vapor, and the second one for directly cooling the vapor. The powder is recovered in a conventional collection unit. The two step condensation in the quench tube allows a substantial control of powder properties like crystallinity, size distribution and mean particle size.

TITLE

Method and transferred arc plasma system for production of fine and ultrafine powders.

5   ## FIELD OF THE INVENTION

The present invention relates to a method for the production of fine and ultrafine powders of various materials such as metals, alloys, ceramics, composites and the like with controlled physical properties. To carry out the method, a novel and flexible transferred arc plasma system providing the ability to control powder

10   properties with a high production rate has been developed. The transferred arc plasma system comprises a transferred arc plasma reactor and a separate quench system within which powder condensation occurs.

BACKGROUND OF THE INVENTION

15   Fine powders of metals, alloys, ceramics, composites and the like have a wide variety of applications in various fields such as aeronautics, electronics, microelectronics, ceramics and medicine. Currently, generation of fine powder, i.e., powders having an average particle size between 0.1 and 10 μm, is mainly accomplished via 3 different techniques: 1) hydrometallurgy, 2) spray pyrolysis and 3)

20   milling. Among the disadvantages of the above techniques are high operating costs, production of non-spherical particles and generation of toxic or difficult to handle by-products.

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The benefits obtained with ultrafine powders, i.e., powders with an average particle size lower than 100 nm, are mainly due to their small particle size, which results in a higher surface area/volume ratio. Consequently, ultrafine powders may have advantages over fine powders when used in the above fields.

5

The preferred methods for the production of fine powders are hydrometallurgy and spray pyrolysis. However, these methods have several major drawbacks including preparation and handling of the feed materials like chlorides and nitrates, which are very often toxic and difficult to handle, environmental emission

10   control requirements for gaseous and liquid effluents, and a difficulty to produce average particle sizes below 100 nm.

Thermal plasma based vapor condensation methods have demonstrated their ability to generate average particle sizes below 100 nm without the handling and

15   environmental problems associated with hydrometallurgical and spray pyrolysis methods. These problems are avoided because the feed materials are generally inert. Examples of such materials include pure metals, alloys, oxides, carbonates etc. Such plasma methods are able to vaporize or decompose these feed materials because of the high-energy input that can be achieved.

20

Thermal plasma generation is typically accomplished via 2 methods, i.e., high intensity DC arcs which uses currents higher than 50 A and pressures higher than 10 kPa, or high frequency discharges such as an RF plasma. Because of their high-energy efficiency, DC arcs are generally preferred. DC arcs are classified as

transferred when one of the electrodes is a material being processed, and non-transferred when the electrodes are non-consumable. Since transferred arc systems pass electrical current directly through a material being processed, their energy efficiency is higher than non-transferred arc systems. Because of the extremely high

5   heat input into the material acting as the electrode, vaporization or decomposition occurs, thus producing a vapor phase that is then cooled to induce the formation of the powder. The powder product is then typically recovered in a filtration unit.

Thermal plasma based vapor condensation methods which utilize a

10   transferred arc have not been successful up to now to generate fine or ultrafine powders of materials like metals, alloys, ceramics or composites on a commercial scale because of their low energy efficiency, low production rate, poor yield, and rudimentary control of powder properties such as particle size and distribution, shape, and crystallinity. In addition, this method is typically used for the production of

15   powders with an average particle size lower than 0.1 μm, which has also contributed to its lack of success on an industrial scale because today's market requires powders with larger particle sizes.

In addition to producing fine and ultrafine powders of various pure

20   materials, transferred arc plasma systems can also be used for the production of fine and ultrafine powders resulting from the interaction of two or more components (chemical reaction) or elements (alloying).

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Although transferred arc plasma systems can operate batchwise, it is preferred that they be operated in a continuous manner. The material to be vaporized or decomposed can be fed continuously in the reactor in several manners. For example, it can be fed into a crucible either from the top thereof by a side tube in the

5   reactor wall. The material can also be pushed upward underneath the plasma in a continuous manner, or fed directly into the plasma torch. Depending on the powder to be produced, the operator will select the appropriate method. Generally, the preferred feeding method is through one or more tubes located in the upper portion of the reactor. The feed materials can be in solid (wire, rod, bar, chunks, shots etc.) or liquid

10   form. When in liquid form, the feed material can also be pumped into the reactor.

U.S. Patent No. 4,376,740 discloses a method for producing fine metal powders which involves reacting a molten metal or alloy with hydrogen using an arc or plasma discharge, or an infrared radiation which dissolves the hydrogen in the

15   metal. When the dissolved hydrogen is released from the molten metal, fine metal powders are generated. Using this method, a low production rate and yield is attained because of the use of a cold-walled reactor and a water-cooled copper mold which is used to support the material being processed. The maximum production rate reported is less than 240 g/hr. Further, there is no mention or suggestion of control of powder

20   properties.

A critical aspect of transferred arc plasma systems is that they consume a lot of energy. It is therefore imperative to maximize its efficiency to have a viable commercial method. This means that the temperature within the reactor must be

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maintained as high as possible to prevent condensation of the vaporized or decomposed materials therein, either on the plasma chamber walls, outside surface of the plasma torch or the mold, which is very often a crucible. Such maximization would obviously result in higher production yields of powders. Because of the

5 extreme conditions prevailing in the transferred arc reactor, many elements are generally water-cooled to extend their operating life. Obviously, such cooling has the effect of reducing the energy efficiency of the method. It has been proposed in Ageorges et al. in *Plasma Chem. and Plasma Processing*, 1993, 13 (4) 613-632 to modify the interior of a transferred arc reactor by covering its internal surfaces with a

10 graphite lining to retain as much heat as possible inside the reactor.

Ageorges et al. *supra*, also disclose the production of ultrafine aluminium nitride (AlN) powder using a transferred arc thermal plasma based vapor condensation method. Vaporizing aluminium and reacting it with nitrogen and

15 ammonia in an insulated plasma chamber produces the desired aluminium nitride product. Aluminium is vaporized by using it as the anode material in a transferred arc configuration that employs a thoriated tungsten tip cathode. The aluminium being vaporized is in the form of an ingot placed in a graphite crucible surrounded by a water-cooled stainless steel support. Because of the presence of that water-cooled

20 jacket, the energy efficiency of vaporization is reduced. A disadvantage of this process is due to the fact that the formation of powder occurs in the plasma chamber because of the injection of reactive gases in the plasma chamber, i.e., nitrogen and ammonia. Ageorges et al. specifically state that the plasma chamber is "filled with fume products which recirculate in the furnace". As a result, powder property control

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is very crude because of the difficulty in properly controlling nucleation and growth of the powder product in the plasma chamber. The particles produced are reported to have a nominal particle size of 135 nm based on specific surface area measurements.

5   To better control the formation of ultrafine aluminium nitride powder, Moura et al. in *J. Am. Ceramic Soc.*, 1997, 80 (9), 2425-2428 propose the separation of aluminium vaporization and aluminium nitride formation. This is accomplished by vaporizing an aluminium anode in a transferred arc reactor in which no reactive gas is introduced, and reacting the aluminium vapor with ammonia injected at a single point 10   in a separate reactor tube attached to the exit of the plasma chamber. The aluminium nitride powders generated with this method have a mean particle size of approximately 20 nm.

15   Da Cruz et al. in *IEEE Trans. on Plasma Science*, 1997, 25 (5), 1008-1016 reports using a thermal plasma based vapor condensation method using a DC transferred arc plasma system. In this work, an aluminium anode is vaporized by striking a thermal Ar or Ar/H_2 arc to it. The aluminium vapor is reacted and cooled rapidly in a separate quench tube to generate ultrafine aluminium nitride powders. The reactor exit gas containing the aluminium vapor is quenched at a single point 20   using an Ar/NH_3 mixture resulting in the production of ultrafine powders. This technique is similar to that described by Moura et al. *supra*. The powders produced have very high specific surface area (40 – 280 m^2/g) and an average particle size of less than 50 nm.

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Chang et al. in *Third Euro-Ceramics*, 1993, 1, 15-20 use a transferred arc thermal plasma based vapor condensation method to produce ultrafine ceramic and composite powders. In their production of SnO_2 or Ag/SnO_2 powders, a tin or silver/tin anode is vaporized by striking an arc to it while it is contained in a graphite

5   crucible surrounded by a water-cooled stainless steel support similar to that described by Ageorges et al. *supra*. A reactive gas, i.e., oxygen, is added to the plasma chamber, resulting in the formation of the product that is then transported to a quenching section. Because oxygen is added into the plasma chamber, both vaporization and reactive steps are conducted in one vessel. In the works of Da Cruz et al. and Moura

10   et al. *supra*, the vaporization and reactive steps in the production of the powder compound are separated to better control the particle formation process.

Chang et al. in *12th International Symposium on Plasma Chemistry*, 1995, 1207-1212 use a similar method to that of Chang et al. in *Third Euro-Ceramics*

15   *supra*. In this work, silica powders are produced. The silica raw material is vaporized by injecting it in a particulate form, i.e., sand with a particle size of 100 – 315 µm, into the arc. The arc is struck between a non-consumable cathode and an anode that is not made of the material being vaporized. As a result, the energy efficiency of this method is likely to be lower than those previously mentioned which use true

20   transferred arc operation. Most of the particles that made up the silica powder product had a particle size ranging from 50 to 400 nm.

It has been shown theoretically that by controlling the initial vapor concentration and temperature, residence time of particle nucleation and growth, and

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cooling profile, one may have some control on the particle size and distribution and crystallinity. This is shown by Okuyama et al. in AIChE Journal, 1986, 32 (12), 2010-2019 and Girshick et al. in *Plasma Chem. and Plasma Processing*, 1989, 9 (3), 355-369. When this method is used for fine powder production as demonstrated by

5 Ageorges et al. and Chang et al. *supra*, control of the powder properties is very crude because no apparatus or procedure is described to accurately control the nucleation, growth and crystallization of particles in the quench section. In addition, in both the works of Ageorges et al. and Chang et al. *supra*, no attempt is made to limit the nucleation and growth of particles in the plasma chamber, which also contributes to

10 the lack of proper control of powder properties. Control of the particle size and distribution, and crystallinity of fine and ultrafine powders produced using a transferred arc thermal plasma based vapor condensation method is therefore very limited.

15   In certain fields, such as electronics or metallurgy, mean size and distribution, and crystallinity of the powder represent critical properties. Accordingly, if such properties can be controlled during the manufacturing process of the powders, it would give to its producer a significant advantage over current fine and ultrafine powder manufacturers.

20

SUMMARY OF THE INVENTION

In accordance with the present invention, there is now provided a transferred arc thermal plasma based vapor condensation method for the production of

fine and ultrafine powders of materials such as metals, alloys, ceramics, composites, and the like. More specifically, the method comprises the steps of:

-   providing a material to be vaporized or decomposed in a plasma reactor;

-   striking an arc between the material and an electrode to generate a plasma having a temperature sufficiently high to vaporize or decompose the material and form a vapor thereof;

-   optionally injecting a diluting gas in the plasma reactor;

-   transporting the vapor by means of the plasma gas and optional diluting gas into a quench tube wherein the vapor is condensed and powder formation occurs, the quench tube comprising

    -   a first section for indirectly cooling or heating the vapor and any particle present therein, to substantially control particle growth and crystallization; and

    -   a second section coupled to the first section for directly cooling the vapor and any particle present therein; and

-   collecting and optionally filtering the powder particles in a collection unit.

In a preferred embodiment, the diluting gas is heated to a temperature corresponding to that of the vapor, or at least 1000 K, before being injected continuously or semi-continuously in the plasma chamber. The injection flow rate of the diluting gas can be varied depending on several parameters such as production rate, powder properties, plasma gas flow rate, vapor concentration etc. Any operator skilled in the art can determine the optimum diluting gas injection flow rate.

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In a further preferred embodiment of the present method, a straight polarity configuration is used, i.e., the liquid material in the crucible is the anode and the electrode is the cathode. In addition, the electrode is non-consumable and is located inside the plasma torch.

5

In a second aspect of the present invention, there is provided a quench tube suitable for the condensation of vapor such as that produced from a transferred arc reactor. More specifically, the quench tube comprises a first section with an elongated substantially tubular body having cooling or heating means around the body for indirectly cooling or heating the vaporized material passing therethrough, thus controlling the growth and crystallization of the particles; and a second section coupled to the first section comprising means for directly cooling the vapor and particles thereof.

10

15

In a preferred embodiment, the second section comprises an extension of the tubular body of the first section, and the direct cooling is done by injecting a cooling fluid directly onto the vapor.

The inner tube diameter and the length of the first section of the quench tube can be varied depending on various parameters, such as powders to be produced, properties desired for these powders, flow rate of the carrier gas, particle size desired, etc. Any experienced engineer or operator skilled in the art may adjust these parameters according to powder properties desired.

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IN THE DRAWINGS

Figure 1 illustrates the components of a typical transferred arc plasma system.

Figure 2 illustrates a transferred arc plasma chamber suitable for the purposes of the present method.

Figure 3 illustrates preferred embodiments of transferred arc configurations suitable for the purposes of the present method.

Figure 4 illustrates a preferred embodiment of a quench tube according to the present invention.

Figure 5 illustrates another embodiment of the quench tube according to the present invention.

Figure 6 illustrates the size distribution of copper particles produced in accordance with the method of the present invention.

Figure 7 illustrates the controlled mean diameter of copper particles produced in accordance with the method of the present invention.

Figure 8 illustrates the crystallinity obtained for copper and nickel powders produced in accordance with the present method.

DETAILED DESCRIPTION OF THE INVENTION

In accordance with the present invention, there is now provided a method for the continuous or batch production of fine and ultrafine powders of materials like metals, alloys, ceramics, composites, and the like, that allows the substantial control of properties of the powders produced. The properties that can be

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substantially controlled are the mean particle size, the size distribution and the crystallinity.

For the purposes of the present invention, the expression "indirect cooling or heating" can be defined as cooling or heating means wherein the coolant or heating does not come in direct contact with the vapor and condensed particles therein, if any, in the vapor exiting the plasma chamber. On the other hand, "direct cooling" is defined as cooling means wherein the coolant is directly contacted with the material's vapor.

The present method comprises striking an arc between an electrode, preferably a non-consumable electrode inside the torch, and a material acting as the other electrode that can be vaporized or decomposed in a straight or reverse polarity configuration. In a straight polarity configuration, the material vaporized or decomposed acts as the anode and the non-consumable electrode acts as the cathode. The material vaporized or decomposed is therefore in a liquid state. As stated above, suitable materials for the method include any electrically conductive material, such as pure metals, alloys, ceramics, composites etc. Examples of metal powders that can be produced include, without being restricted thereto, powders of silver, gold, cadmium, cobalt, copper, iron, nickel, palladium, platinum, rhodium ruthenium, tantalum, titanium, tungsten, zirconium, molybdenum, niobium and the like, as well as alloys thereof. Examples of ceramic powders include, without being restricted thereto, powders of Al_2O_3, TiO_2, SiC, TaC, Si_3N_4, BN etc. Examples of composite or coated

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powders include, without being restricted thereto, powders of SiC/Si, Si_3N_4/Si, NiO/Ni, CuO/Cu etc.

In a continuous method, the material to be vaporized or decomposed can be added continuously or semi-continuously into a crucible that may or may not be electrically conductive at the operating temperature. Typically, the crucible used to contain the material being vaporized or decomposed is electrically conductive so that an auxiliary electrode connection is not required. If an electrically conductive crucible which does not dissolve or react with the material at the operating temperature is not available, a non-electrically conductive crucible which does not have these limitations can be used along with an auxiliary electrode connection. As for the feed material, it can be in any form including solid particles, wire, rod, liquids etc.

Typical plasma torch feed gas flow rates vary depending on the power level and the torch design. Further, when production rates are increased, dilution of the vapor formed from the vaporization or decomposition of the materials in the plasma chamber may be required. Dilution reduces the concentration of the vapor and prevents significant condensation of the vapor, which would lead to the formation of particles in the plasma chamber and hinder the control of powder properties in a separate quench section, as well as reducing the yield. The diluting gas can be added directly to the plasma gas, but this method is usually limited to the maximum operating flow rate of the plasma torch. This is why it is necessary to have additional means for injecting diluting gases in the plasma chamber. Also, such diluting gas must be injected in the plasma chamber at a sufficiently high temperature to minimize

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local quenching which would also lead to particle generation in the plasma chamber. To that effect, at least one gas port is installed to allow additional hot gas to be injected into the plasma chamber. All the gases added into the plasma chamber are selected to minimize reaction with the vapor.

5

Appropriate flow rates for both the torch and the dilution gases can be easily determined by anyone of ordinary skill in the art. These flow rates are dependent on several factors such as production rate, power level, desired particle size etc. As an example, for producing copper or nickel powders of about 0.5 µm at a production rate of 2 kg/h and a power level of from about 50 to about 100 kW, a diluting gas flow rate and a torch gas flow rate of about 1000 ℓ/min and 60 ℓ/min respectively are required. The plasma chamber pressure is preferably maintained between 0.2 – 2.0 atm, and more preferably around 1 atm.

10

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It is important that the reactor be well insulated to maximize energy efficiency and yield, and also to minimize the condensation of the material vaporized or decomposed within the chamber, thus preventing particle formation therein. The vapor is transported from the plasma chamber to a quench tube where the powder particles are grown, and ultimately condensed. The fine or ultrafine powder product can then be collected through any conventional collection/filtration equipment.

20

The method of the present invention provides an energy efficient method for producing fine and ultrafine powders at a production rate of about at least 0.5 kg/h while avoiding the handling and environmental problems associated with

SUBSTITUTE SHEET (RULE 26)

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conventional hydrometallurgical and spray pyrolysis methods. Current transferred arc

systems can only produce at a rate not exceeding 0.2 kg/h, and lack extensive control

of powder properties. The present method permits the relatively simple and cost

effective production of fine and ultrafine powders of materials like pure metals, alloys,

5   ceramics, composites etc. with the ability to substantially control the properties of the

powders.

The invention will now be described by referring to the drawings,

which are provided to illustrate the preferred embodiments of the invention. These

10   drawings shall in no way be considered as limiting the scope of the invention.

Referring to Figure 1, there is provided a plasma system 10, comprising

a transferred arc plasma reactor 12 insulated with an insulating material 13 such as

alumina felt, a quench tube 14, and a powder collection unit 16. Reactor 12 is coupled

15   to a power supply 11, which is itself coupled to a control panel 15. A supply control

unit 19 is also provided for controlling the supply of gases and water in reactor 12. A

heat exchanger 21 may optionally be inserted between unit 16 and quench tube 14 to

further lower the temperature of the powder before collecting it. When the system is

used in a continuous mode, which is almost always the case, a feeder 23 is provided to

20   feed the material inside plasma chamber 17.

Figure 2 illustrates the interior of plasma reactor 12, which comprises a

plasma chamber 17. An arc 18 is struck between an electrode 33, preferably non-

consumable, located inside torch 20 and material 22 contained in a ceramic crucible

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24.   As material **22** is vaporized or decomposed, further material is added continuously or semi-continuously in the crucible, for example through at least one pipe **26**.   A heated diluting gas, preferably argon, helium, hydrogen, nitrogen, ammonia, methane or mixtures thereof, is injected through a pipe **28** into chamber **17** to transport the vapor from chamber **17** into quench tube **14** via at least one exit port **30**, powder condensation occurring inside quench tube **14**.   The powder product exiting the quench tube **14** can be recovered in any suitable solid/gas or solid/liquid separator, such as a particle filtration unit, a scrubber or the like.

When the system is in operation, the energy required for vaporizing or decomposing material **22** is supplied by arc **18** maintained between material **22**, which is partly or completely liquefied in crucible **24**, and electrode **33**. At least one diluting gas is continuously or semi-continuously injected into plasma chamber **17** in addition to the feed gas of plasma torch **20**, this at least one diluting gas being heated to a temperature preferably corresponding to the temperature of the vapor exiting the plasma chamber, or at least higher than 1000 K, to minimize localized condensation of the vapor.

Typically, transferred arc plasma systems, a straight polarity configuration is used in which electrode **33** acts as the cathode and liquid material **22** acts as the anode. However, a reversed polarity configuration is highly advantageous when high operating currents are used because the erosion of electrode **33** is drastically reduced when it acts as the anode.

Preferred arc lengths are from about 2 to 20 cm, but the operator, depending on the material to be produced, can vary the length at will. Pressure inside chamber **17** is preferably maintained between 0.2 – 2.0 atm, the most preferred operating pressure being 0.8 – 1.2 atm.

5

Figure 3 illustrates various alternative transferred arc arrangements inside the plasma chamber. Figure 3A illustrates a preferred crucible arrangement when the latter is conductive. Figures 3B and 3C illustrate configurations wherein an auxiliary electrode **32** is used. Such configuration is suitable when the electrical

10   conductivity of crucible **24** is not efficient, or when the crucible is not conductive at all. It should be noted that an auxiliary electrode may be used even when the crucible is electrically conductive at the operating temperature. The auxiliary electrode connection **32** may or may not be in direct contact with material **22**. The direct contact to material **22** can be either from the top (Figure 3C), bottom or side. When

15   not in direct contact, auxiliary electrode **32** can be a plasma torch, as per Figure 3B, a water-cooled probe or the feed material.

Preferred materials of construction for crucible **24** include high melting point materials such as graphite, carbides such as tantalum carbide, silicon carbide,

20   titanium carbide etc.; oxides such as magnesia, alumina, zirconia etc.; nitrides such as titanium nitride, tantalum nitride, zirconium nitride, boron nitride etc.; borides such as titanium diboride, tantalum diboride, zirconium diboride etc.; as well as refractory metals such as tungsten, tantalum, molybdenum, niobium etc.

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Figure 4 illustrates a preferred embodiment of a quench tube according to the present invention. The vaporized or decomposed material exits chamber 17 in the form of vapor combined with the diluting gas and the plasma gas, and enters into the first section 34 of quench tube 14. First section 34 allows an indirect controlled

5   cooling or heating of the vapor to nucleate the desired product and control the particle growth and crystallization. The indirect heating or cooling can be done using a heating or cooling fluid that is circulated in channel 29 which is formed by the inner surface of an external coaxial tube 36 and the external surface of tube 38. Tube 36 can also be replaced or combined with one or more heating or cooling elements 40 also

10   surrounding tube 38 throughout part or all of its length. The length of section 34 can be varied depending on the size of particles required, the flow rate of the diluting gas, the properties desired for the powders etc. Tube 36 includes at least one inlet 42 and an at least one outlet 44 to allow fluid circulation therein.

15   In the event that a reagent needs to be added to the vapor to produce powders of a product resulting from the reaction between this reagent and the vapor, the reagent may be introduced in the form of a hot reactive gas at one or more points in the first section 34, for example through an inlet 46. Examples of possible reactive gases include nitrogen, hydrogen, ammonia, methane, oxygen, water, air, carbon

20   monoxide or mixtures thereof. The hot reactive gas is also injected at a temperature preferably close to the temperature of the vapor exiting the plasma reactor, or at least higher than 1000 K, to minimize direct cooling of the vapor. Most preferably, the temperature of the injected hot reactive gas is higher than or at least equal to the temperature of the vapor exiting chamber 17. The inner tube of the quench tube

should be constructed from a material that can support the temperature of the vapor exiting the plasma chamber. A preferred material is graphite.

5   To the first section 34 is coupled a second section 50, provided for the direct cooling of the vapor and any powder particles that may have been formed during the passage in first section 34. Direct cooling is performed by injecting a fluid, whether liquid or gaseous, directly onto the vapor and/or powder particles through at least one inlet 52. Preferred gases for direct cooling of the vapor and the powder particles, if any are present, include argon, nitrogen, helium, ammonia, methane, 10   oxygen, air, carbon monoxide, carbon dioxide or mixtures thereof. Preferred liquids include water, methanol, ethanol or mixtures thereof, which are typically injected as a spray.

    The cross-section of the inner tube 38 can be any shape. As an alternative, tube 38 is an annular design with the vapor flowing through the annular 15   gap. This embodiment is illustrated in Figure 5 wherein an elongated body 27 is provided inside tube 38 to form a channel between the inner surface of body 38 and the external surface of body 27.

    The following examples are provided to illustrate the present invention, 20   and should not be construed as limiting its scope. Fine metal powder production was carried out using a transferred arc thermal plasma system as illustrated in Figure 1 comprising a reactor as illustrated in Figure 2, and a quench tube as illustrated in Figure 4. For this purpose, any conventional plasma torch can be used. The crucible can be graphite or any suitable ceramic. The material to be vaporized was fed into the

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crucible in the form of pellets or shots through a port in the upper section of the plasma chamber.

Example 1

Fine copper powders with controlled mean particle size and distribution were produced. In Tests 1 and 2, the control of mean particle size is demonstrated using different quench tube operating conditions. The transferred arc reactor conditions are substantially similar. A mean particle size of 0.78 μm was obtained in Test 1 and 1.74 μm in Test 2.

TABLE 1

Operating conditions and results for Test 1

Operating Parameters	Conditions	Results
Reactor	Plasma gas flow rate = 40 ℓ/min Ar, 20 ℓ/min H_2 Power = 24.5 kw Chamber pressure = 1.1 atm Diluting gas = 85 ℓ/min Ar, T > 1000 K Crucible material = graphite	Vaporization rate = 1.0 kg/h Particle size distribution: 90% less than (d_{90}) = 1.77 μm 50% less than (d_{50}) = 0.78 μm 10% less than (d_{10}) = 0.21 μm (see Figures 6a and 7a) Span =$(d_{90}$- $d_{10})/ d_{50}$= 2.0 XRD count (2θ =43.3°) = 31300
Quench Tube	Length of first section = 10 cm Diameter of the inner tube = 5 cm Indirect cooling gas = 300 ℓ/min Ar Second section direct cooling = 300 ℓ/min N_2	
Collection	Porous metal filter	

TABLE 2

Operating conditions and results for Test 2

Operating Parameters	Conditions	Results
Reactor	Plasma gas = 40 ℓ/min Ar, 20 ℓ/min H_2 Power = 24.5 kw Chamber pressure = 1.1 atm Diluting gas = 85 ℓ/min N_2, T > 1000 K Crucible material = graphite	Vaporization rate = 0.9 kg/h Particle size distribution: 90% less than (d_{90}) = 3.67 μm 50% less than (d_{50}) = 1.74 μm 10% less than (d_{10}) = 0.74 μm (see Figure 6b) Span = $(d_{90} - d_{10})/ d_{50}$ = 1.7 XRD count (2θ = 43.3°) = 30700
Quench Tube	Length of first section = 25 cm Diameter of the inner tube = 5 cm Indirect cooling gas = 200 ℓ/min Ar Second section direct cooling = 300 ℓ/min Ar	
Collection	Porous metal filter	

In Test 3 control of the size distribution is demonstrated. The size distribution was increased in Test 3 compared to Tests 1 and 2 (see Figure 7).

TABLE 3

Operating conditions and results for Test 3

Operating Parameters	Conditions	Results
Reactor	Plasma gas = 40 ℓ/min Ar, 20 ℓ/min H_2 Power = 24.5 kw Chamber pressure = 1.1 atm Diluting gas = 20 ℓ/min Ar, T > 1000 K Crucible material = graphite	Vaporization rate = 0.9 kg/h Particle size distribution: 90% less than (d_{90}) = 2.91 μm 50% less than (d_{50}) = 0.81 μm 10% less than (d_{10}) = 0.25 μm (see Figure 7b) Span = $(d_{90} - d_{10})/ d_{50}$ = 3.3 XRD count (2θ = 43.3°) = 35800
Quench Tube	Length of first section = 10 cm Diameter of the inner tube = 5 cm Indirect cooling gas = 100 ℓ/min Ar Second section direct cooling = 300 ℓ/min N_2	
Collection	Porous metal filter	

Example 2

Fine nickel powders with controlled crystallinity were produced. In Tests 4 and 5 the control of crystallinity is demonstrated using different quench tube operating conditions. The reactor conditions were substantially the same. The degree of crystallinity was measured by the maximum peak count in the X-ray diffraction profile for a given powder sample. This peak occurs at approximately $2\theta = 44.5°$ for nickel and $2\theta = 43.3°$ for copper. The maximum peak count for the nickel powder produced during Test 4 was 24800 compared to 9300 for the nickel powder produced during Test 5.

TABLE 4

Operating conditions and results for Test 4

Operating Parameters	Conditions	Results
Reactor	Plasma gas = 40 ℓ/min Ar, 20 l/min H_2 Power = 28 kw Chamber pressure = 1.1 atm Diluting gas = 65 ℓ/min Ar, T >1000 K Crucible material = graphite	Vaporization rate = 0.5 kg/h Particle size distribution: 90% less than (d_{90}) = 1.42 μm 50% less than (d_{50}) = 0.79 μm 10% less than (d_{10}) = 0.45 μm Span =(d_{90}- d_{10})/ d_{50}= 1.22 XRD count (2θ =44.5°) = 24800 (see Figure 8a)
Quench Tube	Length of first section = 15 cm Diameter of the inner tube = 2.5 cm Indirect cooling gas = 100 ℓ/min Ar Second section direct cooling = 200 ℓ/min Ar	
Collection	Porous metal filter	

TABLE 5

Operating conditions and results for Test 5

Operating Parameters	Conditions	Results
Reactor	Plasma gas = 40 ℓ/min Ar, 20 l/min H_2 Plasma power = 28 kw Chamber pressure = 1.1 atm Diluting gas = 65 ℓ/min Ar, T > 1000 K Crucible material = graphite	Vaporization rate = 0.5 kg/h Particle size distribution: 90% less than (d_{90}) = 1.76 μm 50% less than (d_{50}) = 0.98 μm 10% less than (d_{10}) = 0.54 μm Span =(d_{90}- d_{10})/ d_{50}= 1.24 XRD count (2θ =44.5°) = 9300 (see Figure 8b)
Quench Tube	Length of first section = 15 cm Diameter of the inner tube = 2.5 cm Indirect cooling gas = 300 ℓ/min Ar Second section direct cooling = 200 ℓ/min Ar	
Collection	Porous metal filter	

While the invention has been described in connection with specific embodiments thereof, it will be understood that it is capable of further modifications and this application is intended to cover any variations, uses or adaptations of the invention following, in general, the principles of the invention and including such departures from the present disclosure as come within known or customary practice within the art to which the invention pertains, and as may be applied to the essential features hereinbefore set forth, and as follows in the scope of the appended claims.

WHAT IS CLAIMED IS:

1. A method for the production of fine and ultrafine powders with a transferred arc plasma system, the method comprising the steps of:

- providing a material to be vaporized or decomposed in a plasma reactor;

5 - striking an arc between the material and an electrode to generate a plasma having a temperature sufficiently high to vaporize or decompose the material and form a vapor thereof;

- optionally injecting a diluting gas in the plasma reactor;

- transporting the vapor by means of the plasma gas and optional diluting gas into a

10 quench tube wherein the vapor is condensed and powder formation occurs, the quench tube comprising

- a first section for indirectly cooling or heating the vapor and any particle present therein, to substantially control particle growth and crystallization; and

- a second section coupled to the first section for directly cooling the vapor and

15 any particle present therein; and

- collecting and optionally filtering the powder particles in a collection unit.

2. A method according to claim 1 wherein the quench tube is an elongated substantially tubular body.

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3. A method according to claim 1 wherein the material comprises metals, alloys, ceramics and composites.

4. A method according to claim 1 wherein the material vaporized or decomposed is the anode and the electrode is the cathode and is non-consumable.

5. A method according to claim 1 wherein the material is provided in an electrically conductive crucible.

6. A method according to claim 1 wherein the material is fed in the plasma reactor in the form of solid particles, wire, rod, liquid, or mixtures thereof.

7. A method according to claim 6 wherein the crucible is made of graphite, a carbide, an oxide, a nitride, a boride, or a refractory metal.

8. A method according to claim 2 wherein the indirect cooling or heating is carried out by circulating a cooling or heating fluid in a channel around the body.

9. A method according to claim 1 wherein a reagent is injected in the form of a hot gas into the first section through at least one inlet.

10. A method according to claim 1 wherein the direct cooling is carried out by injecting a cooling fluid directly onto the vapor.

11. A method according to claim 1 wherein the diluting gas is heated at a temperature of at least 1000 K.

12. A method for the production of a fine powder of a metal with a transferred arc plasma system, the method comprising the steps of:

- continuously providing a metal to be vaporized in an electrically conductive crucible inside a plasma reactor;

5 - striking an arc between the metal and a non-consumable electrode in a straight polarity configuration to generate a plasma having a temperature sufficiently high to vaporize the metal and form a vapor thereof;

- injecting a diluting gas heated to a temperature of at least 1000 K in the plasma reactor;

10 - transporting the vapor by means of the plasma gas and the diluting gas into a quench tube wherein the vapor is condensed and powder formation occurs, the quench tube comprising

- a first section comprising an elongated tubular body for indirectly cooling or heating the vapor and any particle present therein, to substantially control
15 particle growth and crystallization; the vapor passing through the inside of the body, and

- a second section coupled to the first section for directly cooling the vapor and any particle present therein by injecting a cooling fluid directly onto the vapor; and

20 - collecting and optionally filtering the powder particles in a collection unit.

13. A method according to claim 12 wherein the metal comprises silver, gold, cadmium, cobalt, copper, iron, molybdenum, nickel, niobium, palladium, platinum, rhodium ruthenium, tantalum, titanium, tungsten, zirconium and alloys thereof.

14. A quench tube for the condensation of a vaporized material, the quench tube comprising an elongated substantially tubular body having a first section comprising cooling or heating means around the body for indirectly cooling or heating 5   the vaporized material passing therethrough to allow controlled growth and crystallization of the material; and a second section coupled to the first section comprising means for directly cooling the vapor and particles thereof.

15. A quench tube according to claim 14 wherein the cooling or heating means 10   comprise a second coaxial substantially tubular body provided around the elongated body and extending throughout the length thereof, thus forming a channel wherein a cooling or heating fluid is circulated.

16. A quench tube according to claim 14 wherein the first section comprises at 15   least one inlet for injecting a hot gas into the vapor.
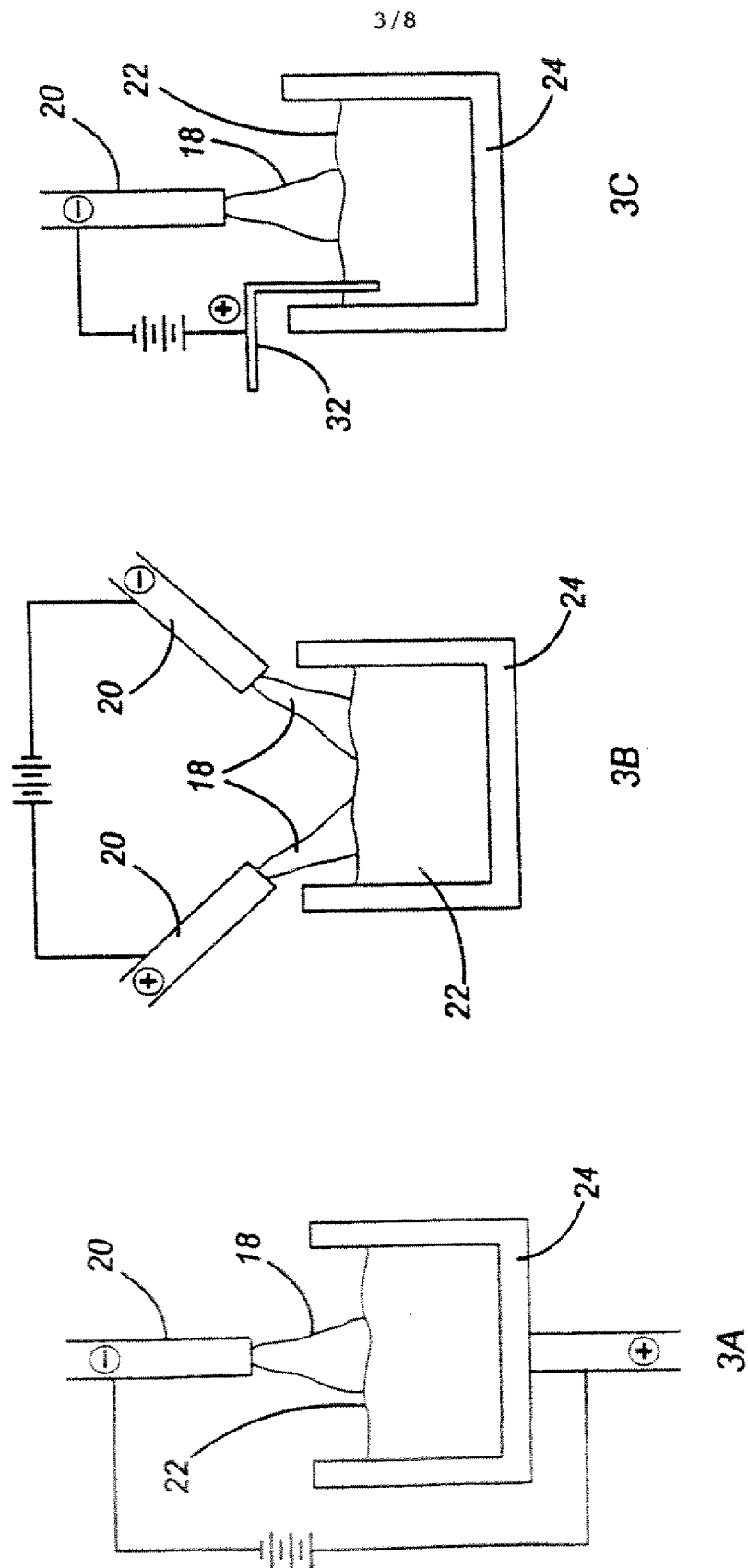
17. A quench tube according to claim 14 wherein the means for directly cooling the vapor in the second section comprises at least one inlet through which a cooling fluid is injected directly onto the vapor.
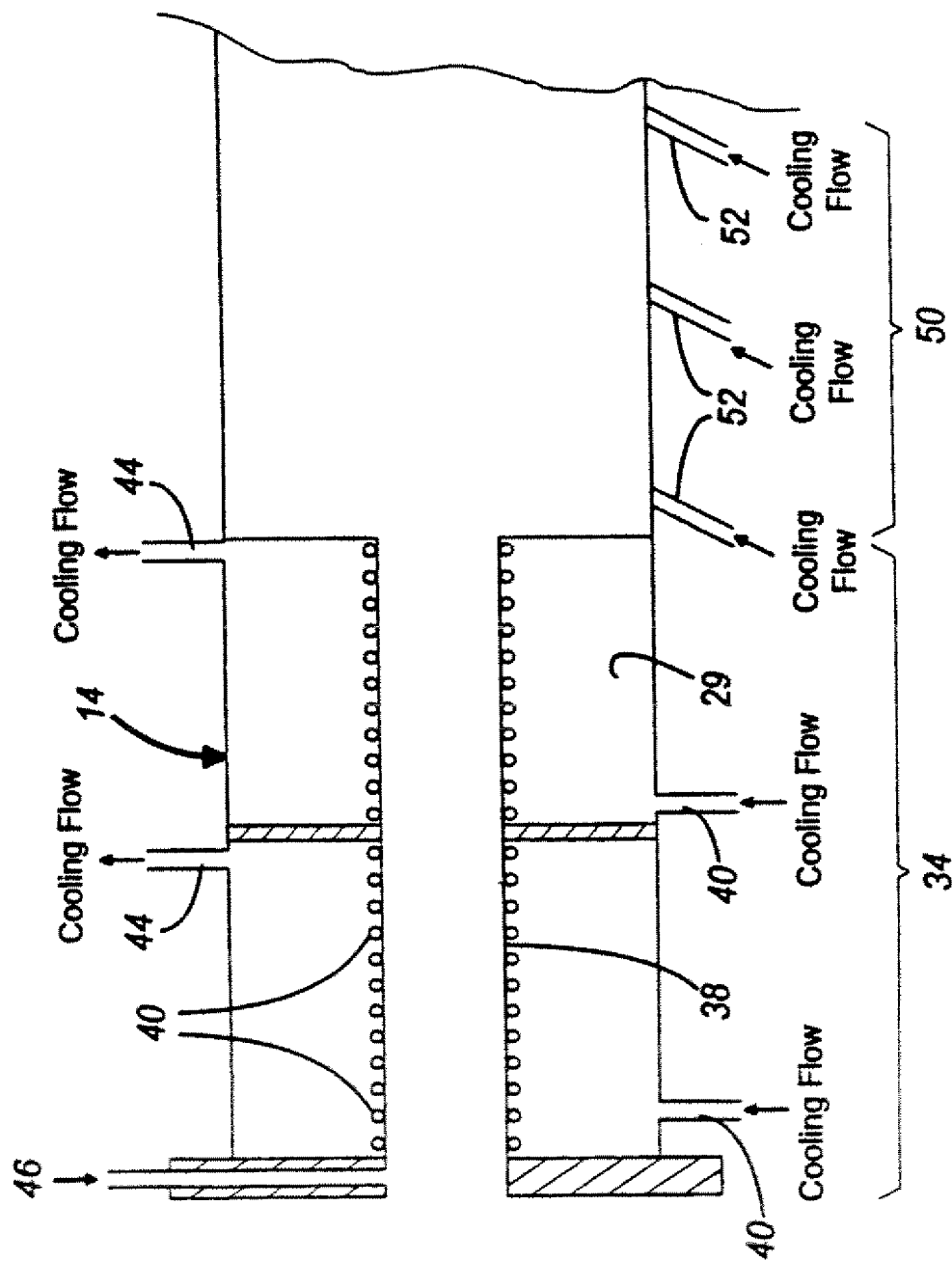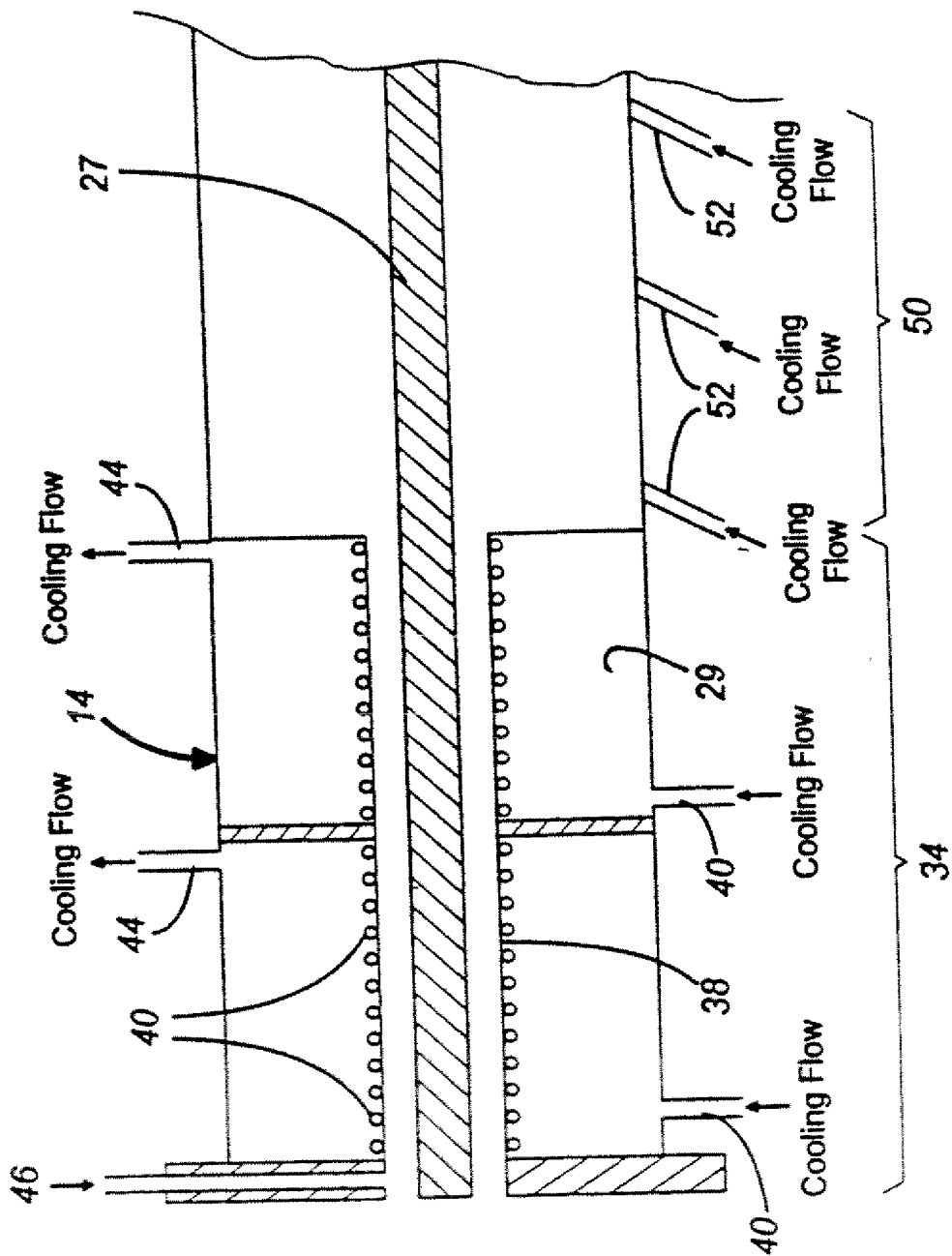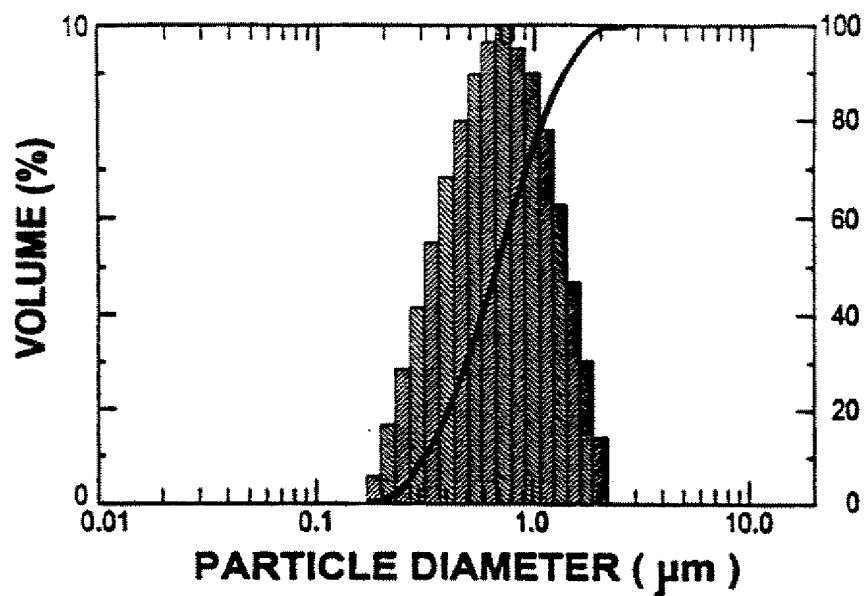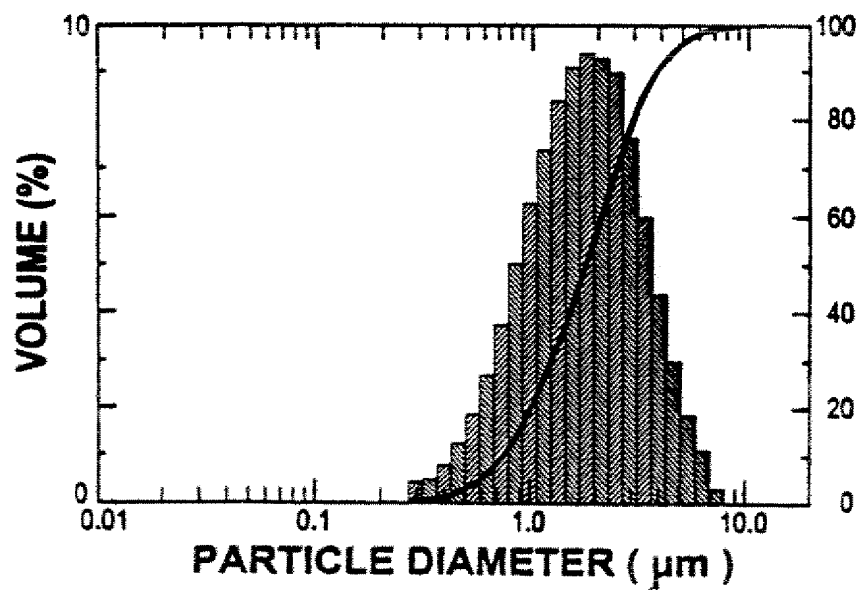
1/8
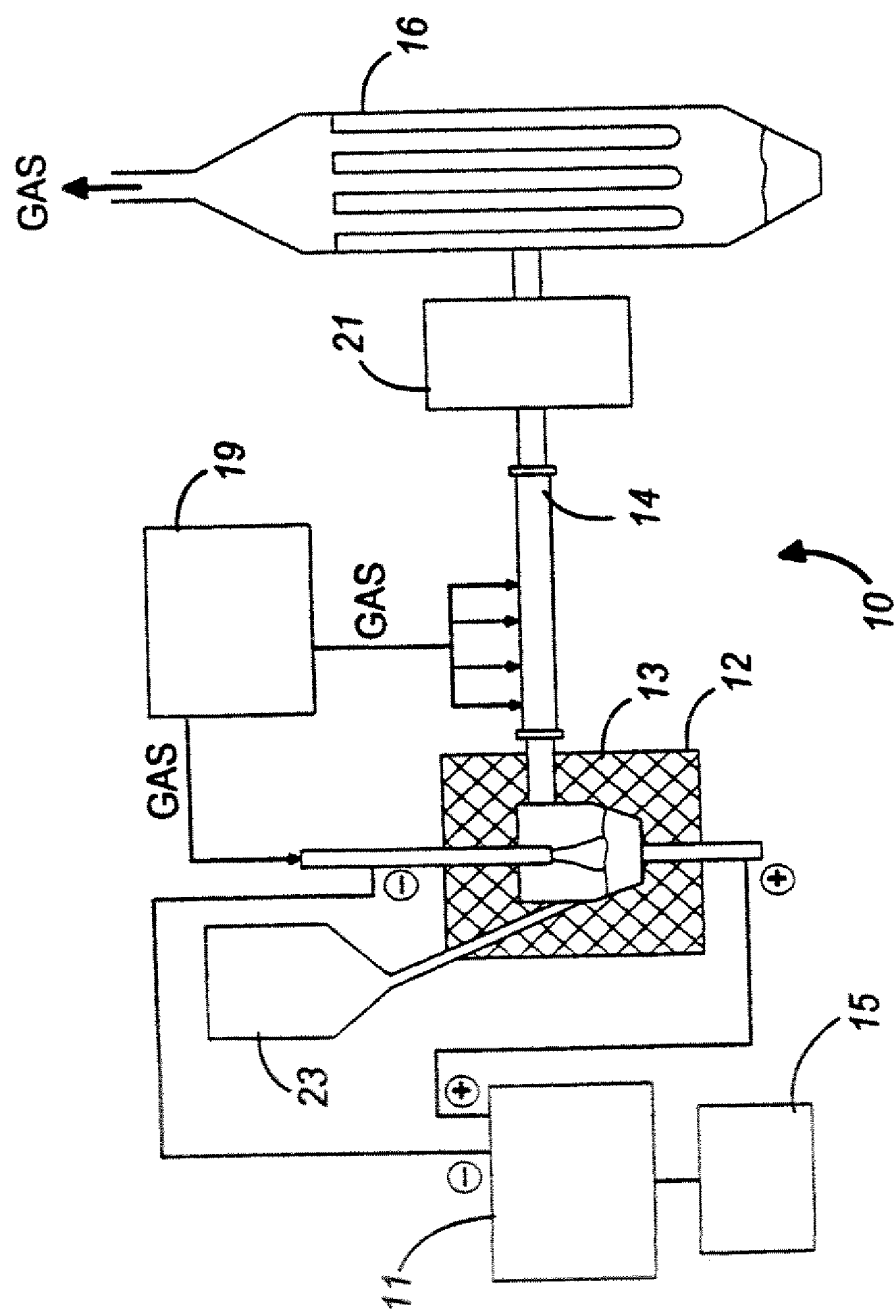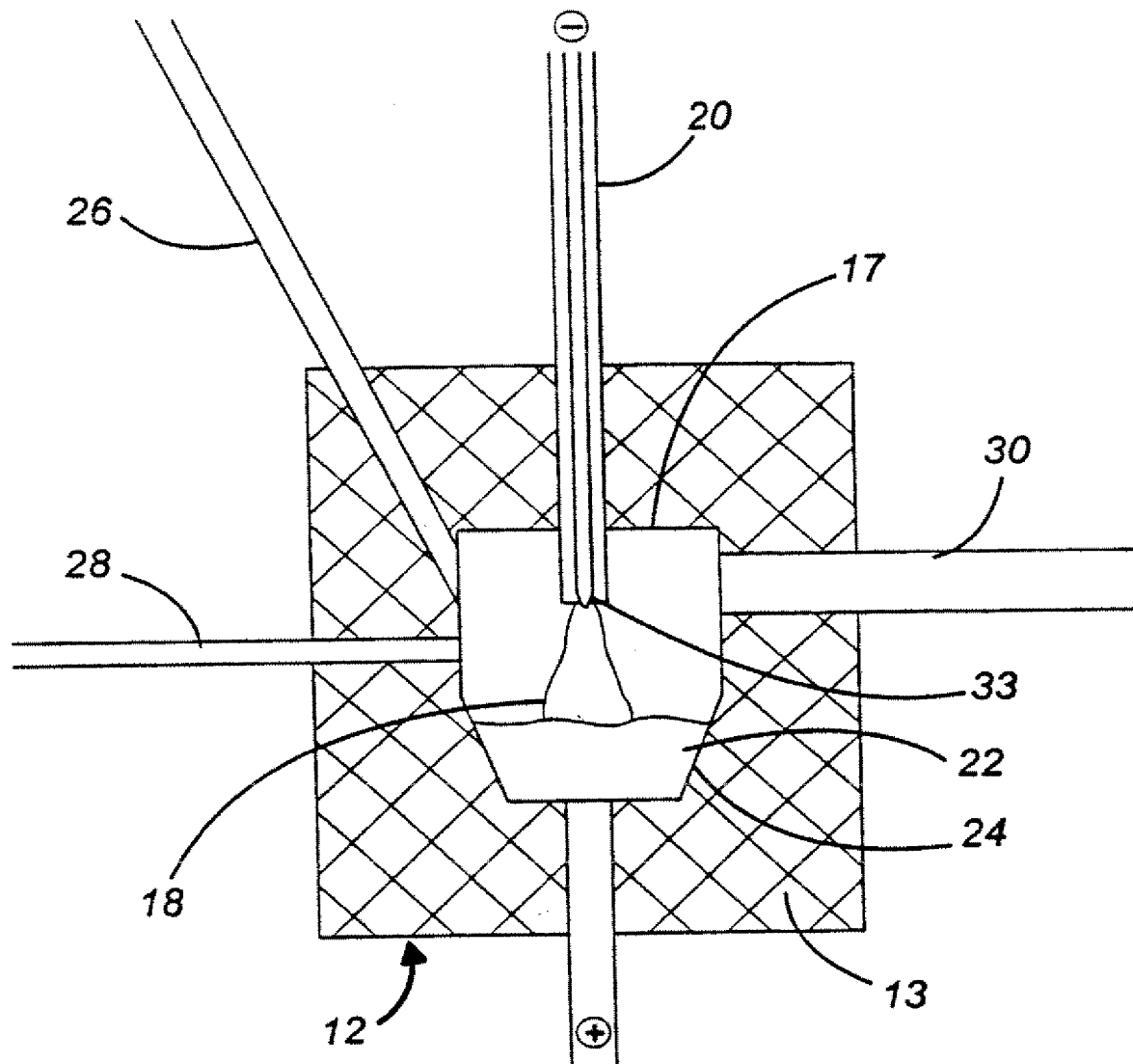
Figure 1

Figure 2

Figure 3

Figure 4

5/8

Figure 5

Cu POWDER

Cu POWDER

Figure 6
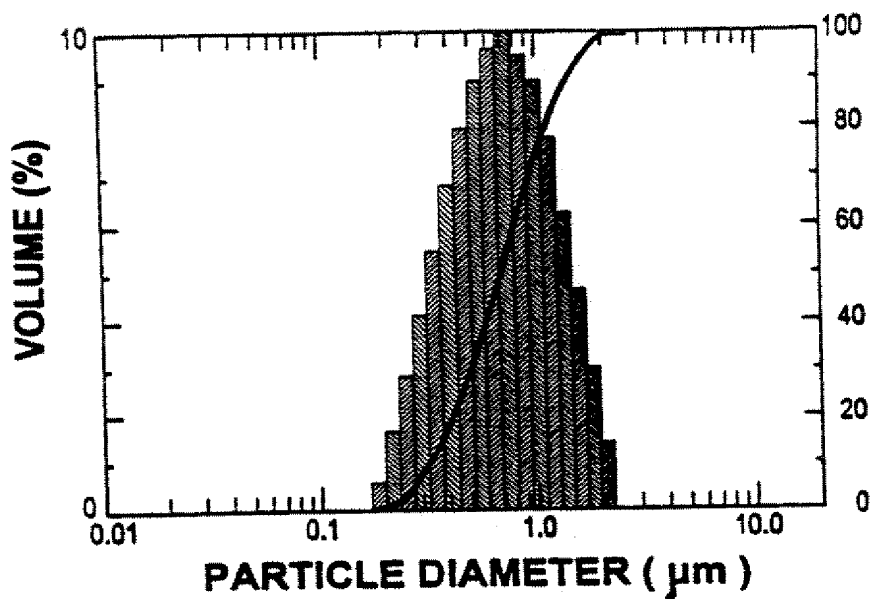
Cu POWDER

Cu POWDER
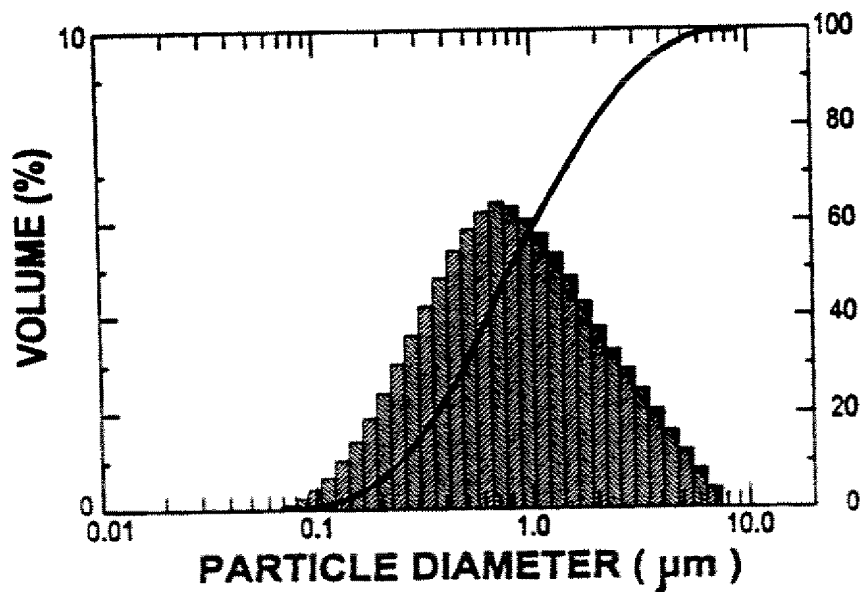
Figure 7
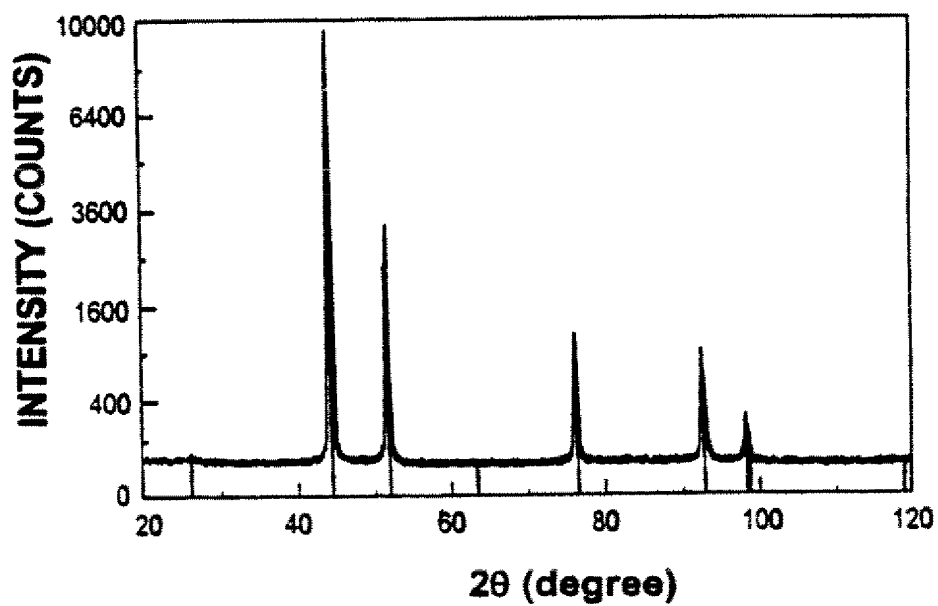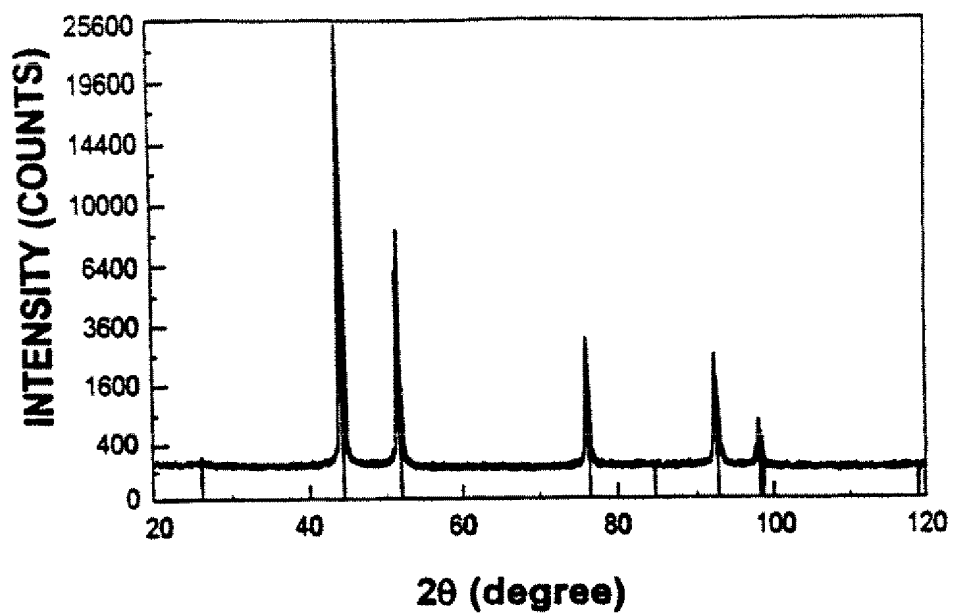
Figure 8

INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7   B22F9/14   B01J19/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7   B22F   C22B   B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	YOUNG R M ET AL: "Generation and behavior of fine particles in thermal plasmas-a review" PLASMA CHEMISTRY AND PLASMA PROCESSING, MARCH 1985, USA, vol. 5, no. 1, pages 1-37, XP002123724 ISSN: 0272-4324 Section 4 page 25, line 18 -page 26, line 2 figure 17 -/--	1

[X] Further documents are listed in the continuation of box C.   [X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such docu- ments, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
24 November 1999	08/12/1999

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Riba Vilanova, M

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INTERNATIONAL SEARCH REPORT

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category°	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
X	RONSHEIM P ET AL: "Direct current arc-plasma synthesis of tungsten carbides" JOURNAL OF MATERIALS SCIENCE, OCT. 1981, UK, vol. 16, no. 10, pages 2665-2674, XP002123725 ISSN: 0022-2461 page 2666, column 1, line 30 -column 22, line 15 figure 1 table 1	1
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A	DD 269 157 A (AKAD WISSENSCHAFTEN DDR) 21 June 1989 (1989-06-21) claim figure	14-17

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